

Mo – 99 FROM LOW – ENRICHED URANIUM

Héctor J. Cols , Pablo R. Cristini , Alberto C. Manzini

Fission Mo-99 Production Plant - Comisión Nacional de Energía Atómica (CNEA)

República Argentina

ABSTRACT

The use of low-enriched uranium aluminide targets for the production of fission Mo-99 is presented. Former tests on uranium silicide targets are also reviewed. Tests of dissolution of aluminide miniplates in alkaline medium were performed, showing interesting results for the replacement of current HEU targets.

INTRODUCTION

As it is widely known, Technetium – 99 m (Tc-99m) is the most important radioisotope in nuclear medicine, because of its physical and chemical properties. Tc-99m is a nuclide produced by radioactive decay of Molybdenum-99 and, as a part of many radiopharmaceuticals, is employed in more than 70% of all medical procedures of diagnostic made with radioisotopes.

In Argentina, fission Mo-99 for high activity Tc-99m generators is being produced since 1985.

The targets employed in current production process are manufactured by the CNEA from highly enriched Uranium (U-235 > 90 %) and irradiated in the RA-3 reactor core during 5 days, with a neutron flux of 1×10^{14} n/cm².sec.

Chemical processing of the targets is carried out following the method developed by Dr. Sameh at KfK in Germany (1,2). Several modifications were performed in order to adapt it to local working conditions. Basically, the radiochemical process consists in separation and purification of Mo-99 by ion exchange chromatography.

As a result of the growing interest in reducing nuclear proliferation concerns, some years ago, CNEA started the development of new low-enriched uranium (LEU) targets for fission Mo-99 production.

CURRENT HEU TARGETS

Current targets have an uranium/aluminum alloy core (“meat”) with an aluminum cladding.

Main features and dimensions of the plates are shown in the following table :

TABLE I

HEU ALUMINIUM ALLOY TARGETS FOR FISSION Mo-99 PRODUCTION	
URANIUM ENRICHMENT	89,4 %
TARGET DIMENSIONS	130 mm x 35 mm x 1,4 mm
“MEAT” DIMENSIONS	115 mm x 30 mm x 0,6 mm
U - 235	1,10 grams
TOTAL URANIUM	1,24 grams
AI AMOUNT IN “CLADDING”	11,8 grams
AI AMOUNT IN “MEAT”	5,0 grams
TOTAL AI AMOUNT	16,8 grams
TOTAL – U DENSITY	0,58 grams / cm³
U – 235 DENSITY	0,53 grams / cm³

URANIUM SILICIDE TARGETS

At the very beginning of the development, uranium silicide was the compound chosen for the “meat” of the new LEU targets, as it allowed, because of U density achievable, to keep a similar amount of U-235 within the same geometry as that of 90 % Uranium targets.

Dissolution of U-silicide miniplates in hot NaOH 3M was tested (3, 4), but the alkaline medium showed not to be enough to achieve complete recovery of Mo-99 in the solution. In order to complete the dissolution, hydrogen peroxide had to be added, forming soluble peruranate.

Under these conditions, it was necessary to separate Uranium from Molybdenum, either by re-precipitation or by other means, such as extraction chromatography or ion exchange. Destruction of peruranate cannot be completed by boiling the solution, so potassium permanganate was added to re-precipitate the uranium. The resulting solution was passed through a column filled with strong anion exchange resin (Dowex 1 or AG-1) as the one employed in current production process. Although the molybdenum was completely retained, it was impossible to recover it from the column using several different counter-ions. The cause of this abnormal fact might be the presence of silicon.

These results implied very important changes to be made in the production method (3,4), so, the decision of trying with another type of compound for the target was taken.

URANIUM ALUMINIDE TARGETS

Uranium aluminide plates employed in the tests had the following features :

TABLE II
LEU ALUMINIDE TARGETS
FOR FISSION Mo-99 PRODUCTION

URANIUM ENRICHMENT	19,7 +/- 0,2%
TARGET DIMENSIONS	130 mm x 35 mm x 1,4 mm
“MEAT” DIMENSIONS	115 mm x 30 mm x 0,7 mm
U - 235	1,4 grams
TOTAL URANIUM	7,0 – 7,1 grams
Al AMOUNT IN “CLADDING”	11,9 grams
Al AMOUNT IN “MEAT”	4,6 grams
TOTAL Al AMOUNT	16,5 grams
TOTAL – U DENSITY	3,0 grams / cm³
U – 235 DENSITY	0,58 grams / cm³

Fabrication of these targets, as well as that of silicide, was carried out by the ECRI group of CNEA, in Constituyentes Atomic Center.

ALKALINE DISSOLUTION TESTS

These test were performed under the same conditions of present process, it is NaOH 3M at 70-80 °C.

COLD TESTS

Plate 7A3 (natural uranium)

This plate was cutted in pieces in order to perform different tests:

- a) Dissolution with NaOH 3M at 70 °C
Cladding dissolved violently in twenty minutes, then the reaction rate slows down to zero at 75 minutes. "Meat" remained undissolved.
- b) It was decided to test dissolution in two stages, first dissolving the cladding and then the "meat" with sodium hypochlorite.
In 20 minutes, cladding was completely dissolved in NaOH 1M at 70 °C. After filtration of the resulting solution, the "meat" was heated with NaClO 17 % w/v at 80 °C, achieving only partial dissolution after 1 hour, then reaction ceases.
- c) It was similar to test b), but with a lower concentration of NaClO (12 % w/v) and three times the original volume. Dissolution of the cladding proceeded as in test b) and meat dissolved completely after 45 minutes.

Plate 8A2 (natural uranium)

Based on the results for 7A3, this plate was dissolved in two stages. First the cladding was completely dissolved in 70 minutes using NaOH 1M at 80 °C. After filtration, NaClO at 90°C was added (600 ml NaClO 2,13 % w/v). Reaction rate was slow, and after two hours the reaction stopped, resulting in partial dissolution of the "meat".

HOT TESTS

Plates MAE 001 and MAE 002 (uranium 20% enriched)

Both plates were irradiated during 108 hours with 1×10^{14} n/*cm².sec neutron flux, and 24 hours of cooling. Plate 002 was kept for post-irradiation examination (PIE). MAE 001 was processed in the hot cells.

Cladding dissolution was performed with 700 ml of NaOH 1M during 75 minutes at 80°C.

The solution was filtered through a sintered stainless steel plate. " Meat" was dissolved with NaClO 12% w/v during 1,5 hours at 75°C. Dissolution was not complete. Excess of NaClO was destroyed adding hydrogen peroxide, because its presence interferes retention of Mo in the first ion exchange column (AG-1). The amount of Mo in loading solution

was only 50 % of the expected, because of partial dissolution of the meat. The resin column didn't worked properly as well, because of excessive oxidant concentration in the solution.

THERMAL TREATMENT

The difficulties found in dissolving the meat of the targets, led to a thorough study of the fabrication process and the U -Al compounds involved (5)

As result of these studies, it was decided to anneal the plates at 485°C in order to form a compound suitable for alkaline dissolution, possibly a mixture of UAl_3 and UAl_4 (5).

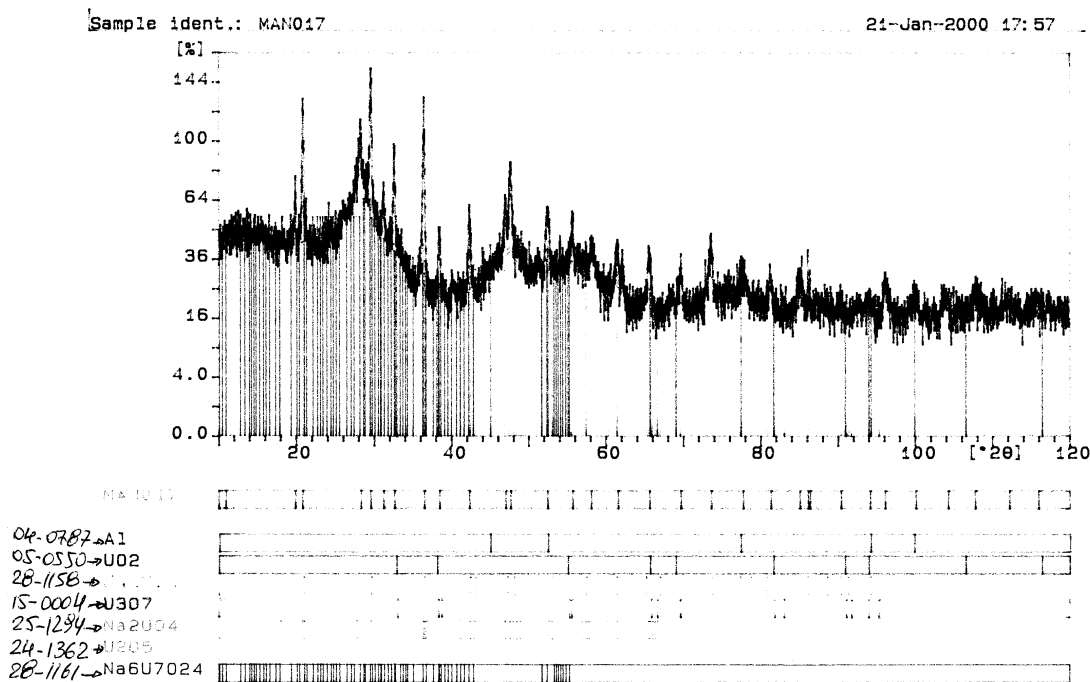
TESTS ON ANNEALED PLATES

Non-irradiated targets

Eight different plates of natural uranium annealed at 485°C were tested . All of them dissolved completely in NaOH 3M at 80°C in 25 – 45 minutes, yielding a black and fine precipitate of uranium dioxide.

Composition of the precipitates was confirmed by X – ray diffraction. One typical X-ray diffraction graphic is shown in the figure.

FIGURE 1



Irradiated targets

Finally, two different tests on irradiated LEU targets were performed. Amount of U-235 was 1,4 grams and total U 7,0 grams per target. They were irradiated during 4 days at 1×10^{14} n/cm².sec with 18 hours of cooling.

Both tests were carried out under identical conditions than that of present method, employing the equipment of routinary production process.

Dissolution was accomplished with NaOH 3 M at 70°C. After two hours, the solution was cooled down to 30°C and filtered using a sintered stainless steel plate. NaClO was added to the solution to oxidize Mo and then, the solution was loaded in a column containing 40 grams of AG-1 x 8 resin at 25 – 30 ml/min. After loading, the column was washed with NaOH 1 M and elution was performed with concentrated sodium sulfate and sodium nitrate solution in ammonium hydroxide 0,5M. Mo recovery from the targets was higher than 90% in both cases.

CONCLUSIONS

The results of testing LEU aluminide targets for fission Mo-99 production were satisfactory. It was proved that annealing of the “meat” at 485°C is needed to get a compound soluble in alkaline medium.

Further experiments has to be performed in order to ensure that the use of LEU aluminide targets will maintain yielding and quality of final product without introducing significant changes in current method. If it is so, uranium aluminide targets will be used for fission Mo-99 production in Argentina in the near future, until the development of a uranium metal foil target will be completed.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the staff of Fission Mo-99 Production Plant for the operation of the hot cells in irradiated targets tests and also the X-ray Diffraction Analysis Group for their work.

REFERENCES

- 1.- R. Marqués, P. Cristini, H. Fernández, D. Marziale “Operation of the installation for fission ⁹⁹Mo production in Argentina” – IAEA – TECDOC – 515 (1989).
- 2.- A. Sameh, H. Ache “Production techniques of fission ⁹⁹Mo” – IAEA-TECDOC – 515 (1989).
- 3.- H. Cols, P. Cristini, R. Marqués “Preliminary investigations on the use of uranium silicide targets for fission Mo-99 production” - RERTR Meeting – Williamsburg, Virginia (1994).
- 4.- H. Cols, R. Marqués, P. Cristini “Research on behaviour of irradiated uranium silicide for fission Mo-99 production” - RERTR Meeting – Paris, France (1995).
- 5.- C. Kohut, M. De la Fuente, P. Echenique “Development of targets of low enrichment for the production of ⁹⁹Mo by fission” RERTR Meeting - Las Vegas, Nevada (2000).